FABRICATION OF EFFICIENT, STABLE AND SCALABLE PEROVSKITE SOLAR CELLS

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FABRICATION OF EFFICIENT, STABLE AND SCALABLE PEROVSKITE SOLAR CELLS

by

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Submitted

in fulfillment of the requirements of the degree of

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to the

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To my parents

and family

CERTIFICATE

This is to certify that the thesis entitled "**Fabrication of Efficient, Stable and Scalable Perovskite Solar Cells**" being submitted by **Mr. Amit Kumar** (Entry No.- 2018ESZ8055) to the Indian Institute of Technology Delhi in fulfillment of the requirements for the award of the degree of **Doctor of Philosophy** is a record of bonafide research work performed by him under my guidance and supervision at Department of Energy Science and Engineering, Indian Institute of Technology Delhi, India. The results obtained herein have not been submitted in part or in full to any other university or institute for the award of any degree to the best of my knowledge.

Date: 10-07-2024

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ABSTRACT

This thesis explores a subset of hybrid perovskite that incorporate formamidinium lead triiodide ($FAPbI_3$) photoactive material for solar cell applications. These perovskites, with a bandgap of 1.52 eV, have shown impressive power conversion efficiencies in solar cells due to their exceptional optoelectronic properties. However, $FAPbI_3$ perovskite exhibits stability challenges. At room temperature, it undergoes a phase change, transitioning from a photoactive black phase (α-phase) to an inactive yellow phase (δ-phase). This phase shift could hinder the long-term performance of the solar cells. To address this, the study presents the synthesis of phase-stable α-$FAPbI_3$ perovskite thin films and solar cells. By incorporating a low concentration of cesium chloride (CsCl), the thin-films obtained exhibits improved phase stability, uniform morphology, and reduced non-radiative recombination. The devices exhibited a power conversion efficiency of ~20% (for an area of 25 mm^2) over 1000 hours, and were briefly discussed in chapter 3. In Chapter 4, we explore how small variations in the chemical composition affect the properties and performance of the perovskite. We find that even a slight excess of methylammonium iodide (MAI) salt can improve phase homogeneity and result in a power conversion efficiency of 19.19% with a larger active area (100 mm^2). Notably, the presence of the PbI_2 phase leads to instability over time, even under inert storage conditions. Additionally, devices degraded rapidly under "dark-85ºC-inert conditions (ISOS-D2 protocol)", when fabricated with n-i-p configuration. To make these findings practical for large-scale applications, we optimize the choice of solvents used in the manufacturing process, as discussed in Chapter 5. Our efforts lead to an impressive power conversion efficiency of 18.66% for a 25 mm^2 area using 2-methoxyethanol (2-ME) solvent system and antisolvent free processing. Chapter 6 delves deeper into the improved stability of 2-ME-processed thin films under thermal stress compared to standard DMF:DMSO films. Additionally, optoelectronic measurements indicate no degradation in 2-ME-processed films under ISOS protocol conditions. Lastly, we identify issues at the interface between the perovskite and C60 (ETL) interface, which limits the overall device performance, through half-stack PLQE measurements. In summary, this thesis strives to produce uniform and stable $FAPbI_3$ perovskite thin films and photovoltaic devices, exploring non-stoichiometry's impact, optimizing a scalable solvent system, and assess their stability under challenging conditions following industry-standard protocols (ISOS-D2 and ISOS-L3 protocols).

सारांश

यह थीसिस हाइब्रिड पेरोव्स्काइट्स के एक उपसमूह की खोज करती है जिसमें सोलर सेल अनुप्रयोगों के लिए फॉर्ममिडिनियम लेड ट्राईआयोडाइड ($FAPbI_3$) फोटोएक्टिव सामग्री शामिल होती है। 1.52 eV के बैंडगैप वाले इन पेरोव्स्काइट्स ने अपने असाधारण ऑप्टोइलेक्ट्रॉनिक गुणों के कारण सोलर सेल में प्रभावशाली बिजली रूपांतरण क्षमता दिखाई है। हालाँकि, $FAPbI_3$ पेरोव्स्काइट में स्थिरता संबंधी समस्याएँ हैं। कमरे के तापमान पर, यह सक्रिय, काले चरण (α- चरण) से निष्क्रिय, पीले चरण (δ- चरण) में एक चरण परिवर्तन से गुजरता है। यह चरण परिवर्तन सोलर सेल के दीर्घकालिक प्रदर्शन में बाधा उत्पन्न कर सकता है। इसे संबोधित करने के लिए, अध्ययन स्थिर α-$FAPbI_3$ पेरोव्स्काइट पतली फिल्मों और सोलर सेल के संश्लेषण को प्रस्तुत करता है। सीज़ियम क्लोराइड (CsCl) की कम सांद्रता को शामिल करके, परिणामी फिल्में बेहतर चरण स्थिरता, समान आकारिकी और कम गैर-विकिरणीय पुनर्संयोजन प्रदर्शित करती हैं। उपकरणों ने 1000 घंटों में ~20% (25 मिमी वर्ग के क्षेत्र के लिए) की बिजली रूपांतरण दक्षता प्रदर्शित की, और अध्याय 3 में संक्षेप में चर्चा की गई है। अध्याय 4 में, हम पता लगाते हैं कि रासायनिक संरचना में छोटे बदलाव पेरोव्स्काइट के गुणों और प्रदर्शन को कैसे प्रभावित करते हैं। हमने पाया कि मिथाइलमोनियम आयोडाइड (MAI) नमक की थोड़ी सी भी अधिकता चरण समरूपता में सुधार कर सकती है और बड़े सक्रिय क्षेत्र (100 मिमी वर्ग) के साथ 19.19% की बिजली रूपांतरण दक्षता में परिणाम कर सकती है। विशेष रूप से, PbI_2 चरण की उपस्थिति समय के साथ अस्थिरता की ओर ले जाती है, यहां तक कि निष्क्रिय स्थितियों में भी। इसके अतिरिक्त, n-i-p कॉन्फ़िगरेशन के साथ निर्मित होने पर, डिवाइस "डार्क-85°C- निष्क्रिय स्थितियों (ISOS-D2 प्रोटोकॉल)" के तहत तेजी से खराब हो जाते हैं। इन निष्कर्षों को बड़े पैमाने पर अनुप्रयोगों के लिए व्यावहारिक बनाने के लिए, हम विनिर्माण प्रक्रिया में उपयोग किए जाने वाले सॉल्वैंट्स की पसंद को अनुकूलित करते हैं, जैसा कि अध्याय 5 में चर्चा की गई है। हमारे प्रयासों से 2-मेथॉक्सीएथेनॉल का उपयोग करके 25 मिमी वर्ग क्षेत्र के लिए 18.66% की प्रभावशाली बिजली रूपांतरण दक्षता प्राप्त होती है। (2-ME) विलायक प्रणाली। अध्याय 6 मानक DMF:DMSO फिल्मों की तुलना में थर्मल तनाव के तहत 2-ME-संसाधित पतली फिल्मों की बेहतर स्थिरता पर गहराई से प्रकाश डालता है। इसके अतिरिक्त, ऑप्टोइलेक्ट्रॉनिक माप ISOS प्रोटोकॉल शर्तों के तहत 2-ME-संसाधित फिल्मों में कोई गिरावट नहीं दर्शाते हैं। अंत में, हम आधे-स्टैक PLQE माप के माध्यम से पेरोव्स्काइट और C60 (ETL) इंटरफेस के बीच इंटरफेस में मुद्दों की पहचान करते हैं, जो समग्र डिवाइस प्रदर्शन को सीमित करते हैं। संक्षेप में, यह थीसिस एक समान और स्थिर $FAPbI_3$ पेरोव्स्काइट पतली फिल्मों और फोटोवोल्टिक उपकरणों का उत्पादन करने, गैर-स्टोइकोमेट्री के प्रभाव की खोज करने, एक स्केलेबल विलायक प्रणाली को अनुकूलित करने और उद्योग-मानक प्रोटोकॉल (ISOS-D2 और ISOS- L3 प्रोटोकॉल) के बाद चुनौतीपूर्ण परिस्थितियों में उनकी स्थिरता का आकलन करने का प्रयास करती है।

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LIST OF ABBREVIATIONS

2ME	2-methoxyethanol
AM	Air Mass
BSE	Backscattered Electron
CBM	Conduction Band Minimum
CE	Counter Electrode
CdTe	Cadmium Telluride
CIGS	Copper Indium Gallium Selenide
CPD	Contact Potential Difference
CSL	Charge Selective Layer
CZTS	Copper Zinc Tin Sulfide
DEF	Defect Formation Energy
DMF	Dimethylformamide
DMPU	Dimethylpropyleneurea
DMSO	Dimethyl sulfoxide
DSSC	Dye-sensitized solar cell
EDX	Energy Dispersive X-ray
ETL	Electron Transport Layer
FF	Fill Factor
FTO	Fluorine doped tin oxide
GaAs	Gallium arsenide
GB	Grain Boundary
GGA	Generalized Gradient Approximation
HOMO	Highest Occupied Molecular Orbital
HTL	Hole Transporting Layer
IQE/EQE	Internal/External Quantum Efficiency
ISOS	International Organization for Standardization
KPFM	Kelvin Probe Force Microscope
LED	Light Emitting Diode
LUMO	Lowest Unoccupied Molecular Orbital
MHP	Metal Halide Perovskite
MPPT	Maximum Power Point Tracking
NAMD	Non-Adiabatic Molecular Dynamics
NMP	N-Methyl-2-pyrrolidone
NREL	National Renewable Energy Laboratory

NRR	Non-Radiative Recombination
PLQE	Photoluminescence Quantum Efficiency
PSC	Perovskite Solar Cells
PV	Photovoltaics
QFLS	Quasi Fermi Level Splitting
QFEL	Quasi Fermi Energy Level
SE	Secondary Electron
SEM	Scanning Electron Microscope
Si	Silicon
SOC	Spin Orbital Coupling
SPO	Stabilized Power Output
SQ	Shockley Queisser
SRH	Shockley-Read-Hall
SSPL	Steady State Photoluminescence
TCSPC	Time Correlated Single Photon Counting
TRPL	Time Resolved Photoluminescence
TW	Terra Watt
VASP	Vienna Ab initio Simulation Package
VBM	Valence Band Maximum
XRD	X-Ray Diffraction